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Actinide Sorption in Rainier Mesa Tunnel Waters

From the Nevada Test Site

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Abstract

The sorption behavior of americium (Am), plutonium (Pu), neptunium (Np), and uranium (U) in perched Rainier Mesa tunnel water was investigated. Both volcanic zeolitized tuff samples and groundwater samples were collected from Rainier Mesa, Nevada Test Site, NV for a series of batch sorption experiments. Sorption in groundwater with and without the presence of dissolved organic matter (DOM) was investigated.

Am(III) and Pu(IV) are more soluble in groundwater that has high concentrations of DOM. The sorption K_d for Am(III) and Pu(IV) on volcanic zeolitized tuff was up to two orders of magnitude lower in samples with high DOM (15 to 19 mg C/L) compared to samples with DOM removed (< 0.4 mg C/L) or samples with naturally low DOM (0.2 mg C/L). In contrast, Np(V) and U(VI) sorption to zeolitized tuff was much less affected by the presence of DOM. The Np(V) and U(VI) sorption K_ds were low under all conditions. Importantly, the DOM was not found to significantly sorb to the zeolitized tuff during these experiment.

The concentration of DOM in groundwater affects the transport behavior of actinides in the subsurface. The mobility of Am(III) and Pu(IV) is significantly higher in groundwater with elevated levels of DOM resulting in potentially enhanced transport. To accurately model the transport behavior of actinides in groundwater at Rainier Mesa, the low actinide K_d values measured in groundwater with high DOM concentrations must be incorporated in predictive transport models.

1. Introduction

Over 800 underground nuclear tests were detonated from 1951-1992 at the Nevada Test Site as part of the United States nuclear testing program (Figure 1). Residual radionuclides were deposited in the subsurface after each nuclear test. This inventory of radioactivity in the subsurface comprises the radiologic source term (RST). The amount of radioactivity that is available for transport in groundwater is referred to as the hydrologic source term (HST). Transport of the residual radionuclide inventory as part of the HST is a function of the initial radiologic source term, geochemistry and hydrology of the subsurface. In the Rainier Mesa and Shoshone Mountain area of the NTS, nuclear tests were emplaced

primarily in an extensive and complex system of tunnels mined into the side of Rainier Mountain within the vadose zone. The perched water in the tunnel complex at Rainier Mesa has a much higher dissolved organic matter (DOM) content than local spring water as a result of anthropogenic activities (tunnel lagging and wood debris, drilling fluids, diesel fuel, etc.) associated with underground nuclear testing and construction of the tunnels. To model the transport behavior of the radionuclides and predict the HST in the Rainier Mesa and Shoshone Mountain region, the effect of groundwater with a high concentration of DOM needs to be evaluated.

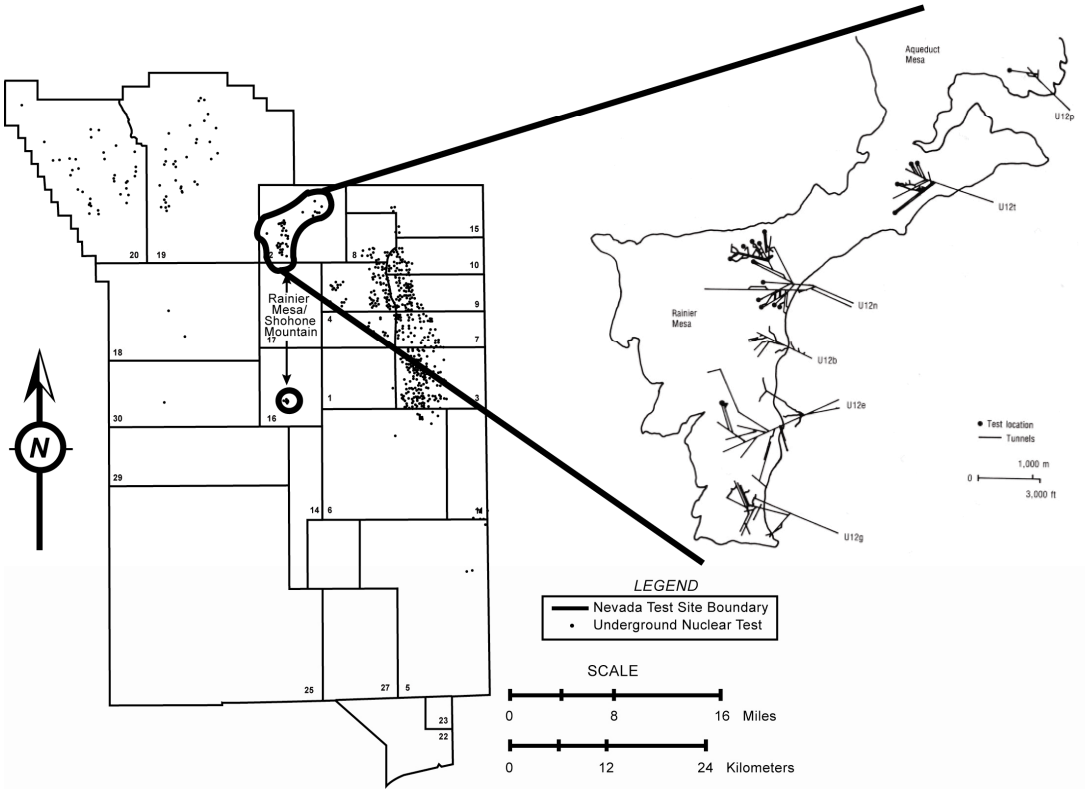

Figure 1. The Nevada Test Site, located 100 km northwest of Las Vegas, NV is outlined on the left. The tunnel system in Rainier Mesa is expanded on the right, showing the location of U12n Tunnel and U12t Tunnel.

The migration of actinides in natural aquatic systems is influenced by many factors including oxidation state (Eh), hydrolysis (pH), sorption, formation of colloids, and colloid filtration (e.g., Lieser et al., 1991, Choppin, 2006). Studies have shown that in organic-rich systems, DOM can significantly facilitate the mobilization and transport of actinides (McCarthy and Czerwinski et al., 1998, Maes and Wang et al., 2006, Mibus and Sachs et al.,

2007, Wolff-Boenisch and Traina, 2006, Vandenhove et al., 2007). DOM in groundwater is composed of a wide variety of compounds including humic substances, hydrophilic acids, carbohydrates, carboxylic acids, amino acids and hydrocarbons (Thurman, 1986). It is known that organic-metal complexation can enhance the solubility of metal ions, therefore, increasing metal concentration in water (Marquardt and Kim, 1998, Alliot et al., 2005, Kantar and Honeyman, 2005).

DOM-metal complexes may also sorb to a given mineral, which is either mobile (i.e. colloids or sediments) or immobile (host rock), thereby increasing or reducing the mobility of the complexes (Plater et al., 1992). Organic matter may sorb on the mineral surfaces to compete with soluble actinides species for sorption sites, hence reducing sorption of the actinides. Furthermore, organic substances can be involved in redox reactions with actinides, resulting in either reduced or oxidized actinide species, which may be less or more mobile in water (Choppin, 1988). The selectivity in the complexation of actinides by humic acids, a component of DOM, may also play a role in either the mobilization or retardation of the actinides (Zhang et al, 1997). Some studies have shown that humic substances have the ability to immobilize and retard the transport of uranium and plutonium (Artinger and Rabung et al., 2002, Choppin, 2006, Reiller et al., 2002). The effect of DOM on the mobility of actinides is not easily predicted, but is a function of the specific actinide, its oxidation state, and the nature of the organic material in the system. Am(III) and Pu(IV) can form complexes with natural organic ligands (Tits et al., 2005). Organic ligands originating from the degradation of cellulose materials can reduce the retardation of tri- and tetravalent actinides because of the formation of the metal-ligand complexes in aqueous phase. Maes et al. (2006) reported the total concentration of Am(III) increases several orders of magnitude above the solubility limit in the presence of soluble organic matter. Nakayama and Nelson (1988) also reported that K_d values for Am and Cm decreased two orders of magnitude when concentrations of colloidal organic carbon increased ~1000 times in natural water.

In contrast to Am(III) and Pu(IV), Np(V) and U(VI) are less likely to form stable complexes with organic carbon (Kung and Triay, 1994, Murphy et al., 1999, Li and Tao, 2003, Khasanova et al., 2007). Niitsu et al. (1997) reported that K_d values for Np(V) on kaolinite increased slightly with an increasing concentration of humic acid when the pH was less than eight and decreased no more than an order of magnitude with higher concentration of humic acid with pH values greater than eight. They suggested that the sorption/desorption of the humic acids on/from minerals played a major role in the sorption behavior of Np(V). Similar results for U(VI) sorption to hematite in the presence of DOM were reported by Lenhart and Honeyman (1999). However, Baston et al. (1994) reported that in the presence of high concentrations of organic degradation products at neutral pH, significant reduction in uranium sorption was observed and K_d values of U(VI) were two orders of magnitude lower than those at pH 12. Plater et al. (1992) reported that the complexation of uranium with DOM in mobile river sediments appeared to increase the mobilization of uranium.

The objective of this study was to compare the sorption behavior of several actinides (Am, Pu, Np, and U) on volcanic zeolitized tuff in groundwater collected from Rainier Mesa where the DOM concentration varies significantly. Four zeolitized tuff samples from the tunnel system in Rainier Mesa were used in the study (Fig. 2). Water samples were collected

from two tunnels, U12t Tunnel and U12n Tunnel. An additional groundwater sample from well ER-12-3 near the tunnel system, that has low level of DOM, was also used.

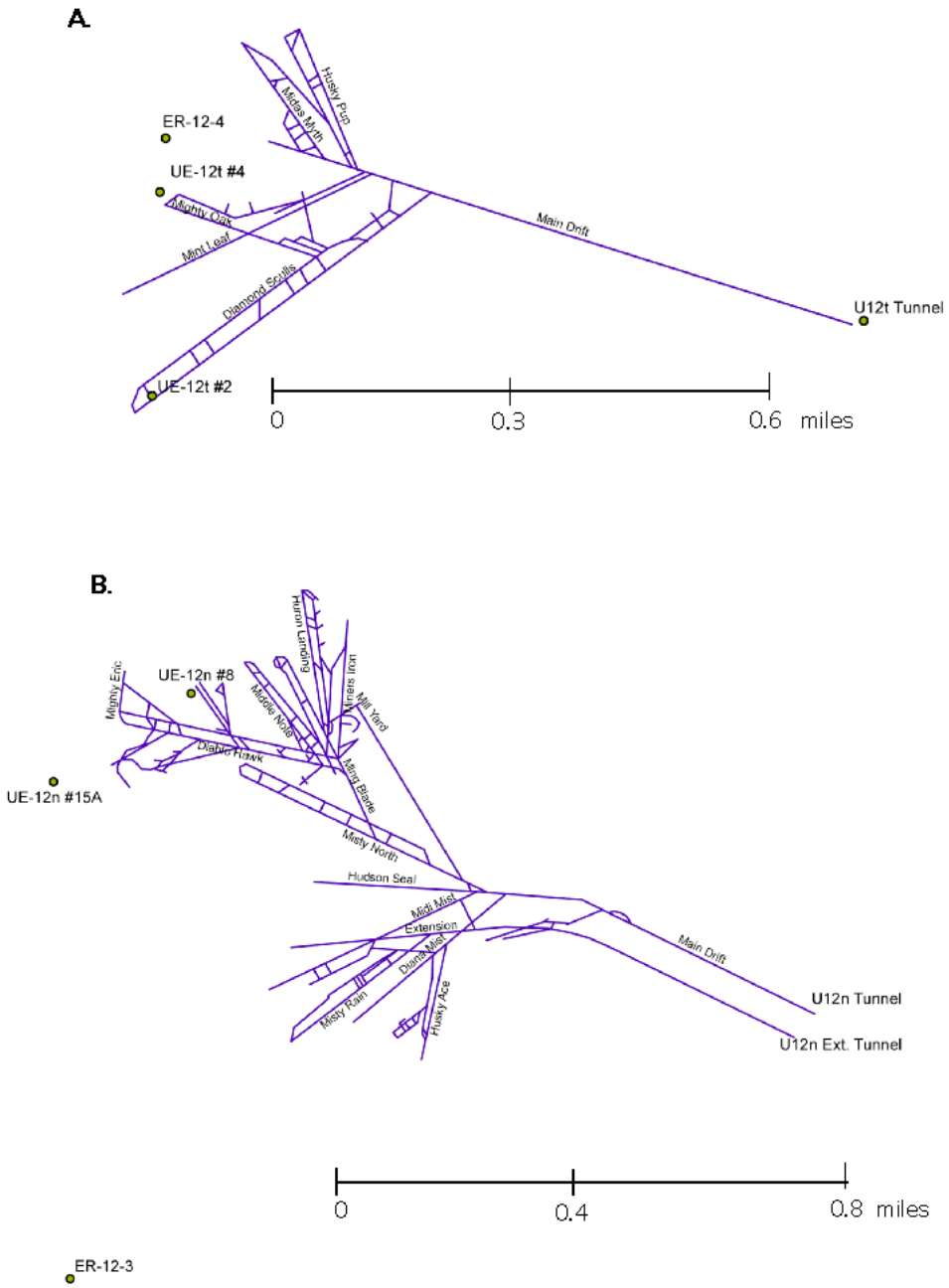

Figure 2. A) Map of U12t Tunnel and B) Map of U12n Tunnel, showing the location of each borehole. ER-12-3 is located southwest to the U12n tunnel complex.

2. Material and Methods

2.1 Reagents

Unless stated otherwise, chemicals used were reagent grade or better. TEVA, TRU and U-TEVA resins (Eichrom Technologies Inc.) were used in the Pu, Am and U purification. The anion exchange resin AG1×8 (100-200 μm mesh size) and poly-prep plastic columns (Bio-Rad Laboratories) were used in actinides purification as well.

2.2 Volcanic zeolitized tuff

Four volcanic zeolitized tuff samples, which were collected from exploratory holes located near the underground nuclear tests in U-12t Tunnel and U-12n Tunnel, were obtained from the NTS core library. The sampling locations, UE-12t #2, UE-12t #4, UE-12n #8 and UE-12n #15A, are indicated on Figure 2 A&B. The zeolitized tuff samples were crushed to a particle size of 500 microns or smaller, then dry sieved to collect the size fractions from 75 to 500 microns. This fraction, which contained greater than 70% of the total crushed mass of each tuff rock, was used in the sorption experiments. The surface area of the samples was measured using Micrometrics Gemini 2370 BET surface area analyzer. The mineral phases were analyzed using x-ray diffraction on a Scintag PAD-V diffractometer.

2.3 Preparation of water samples from Rainier Mesa tunnel complex

Water samples from two different tunnel systems (U12t and U12n) were used in the sorption experiments (collected October 30 to November 2, 2006). Groundwater (low DOM) from well ER-12-3 (collected July 6, 2005) was used in the sorption experiments as well (Figure 2). The water samples were filtered through a 20 nm membrane filter to remove majority of particulates, colloids and microbial matter. Ion chromatography (Dionex) was used to analyze major anions and inductively coupled plasma mass-spectrometry (ICP-MS) was used for major cations. A total organic carbon (TOC) analyzer (Aurora model 1030 from OI Analytical) was used for organic and inorganic carbon analyses.

To examine the effects of DOM on sorption, the two tunnel water samples were treated with activated charcoal (Othman et al., 2000 and 2001) to remove organic matter. The first batch of the tunnel water samples treated with activated charcoal resulted in high concentrations of sulfate. These waters were used for Am & Pu sorption experiments. The method for removing organic carbon was later improved by an additional step designed to lower the sulfate concentration. The additional step involved flashing the charcoal columns with 1.5 liters of 10^{-3} M $NaHCO_3$ solution prior to pumping the tunnel waters through the charcoal columns. The water prepared by this 2-step process had much lower sulfate concentrations and was used for U and Np sorption experiments. A subsequent series of Am and Pu sorption experiments were carried out using low sulfate waters to determine if the increase in sulfate resulting from the removal of organic carbon affected the sorption of Am and Pu to the tuff. The increase in sulfate concentration did not appear to affect the K_d of Am or Pu to the zeolitized tuff.

The dissolved organic compounds in the tunnel water samples were also extracted for characterization purposes using solid phase extraction (SPE) cartridges (Supelco supelclean

ENVI-18, 6 mL, 0.5 g). A liter of each water sample was pumped through the SPE cartridges resulting in brown discoloration in the cartridges. The organic material collected in the SPE cartridge was first air-dried and then eluted using methanol /dichloromethane. The SPE cartridges remained brown in color after the elution suggesting that some organic material may have remained adsorbed to the cartridge material. The organic extract was then analyzed by gas chromatography-mass spectrometry (GC-MS).

2.4 Radionuclides

Alpha emitters ^{241}Am, ^{238}Pu, ^{233}U and ^{237}Np were used in the batch sorption experiments. The ^{241}Am in 3M HNO_3 was purified using TEVA and TRU resin columns. The purified ^{241}Am was eluted using 1M HCl solution. ^{238}Pu in 4M HNO_3 was purified using a TEVA column and eluted using a 1M HCl as a final stock solution. The oxidation state of the ^{238}Pu stock solution was characterized using both solvent extraction with PMBP (1-phenyl-3-methyl-4-benzoyl-5-pyrazolone) and Pu co-precipitation by lanthanum fluoride. The results indicated that the Pu stock solution consisted of 80% Pu(IV), 15% Pu(III) and 5% colloidal Pu. The ^{237}Np stock was purified in concentrated HCl with KI solid using an AG1x8 (100-200 mesh) resin column. The Np was eluted from the column using 0.1M HNO_3. The Np solution was dried in HNO_3 on a hotplate before re-dissolving in 1M HCl to make a Np(V) solution. Np(V) oxidation state of the stock solution was confirmed by UV/VIS spectrum. The ^{233}U stock in 4M HNO_3 solution was purified by a UTEVA column and eluted from a column using 0.1M HCl. The oxidation state of U in the final stock solution was U(VI). The activities of all four radioisotope stock solutions were determined using a liquid scintillation analyzer. The purity of all four radioisotope stock solutions (~100% pure in activity) was confirmed using alpha spectrometry.

2.5 Batch sorption experiments

All batch sorption experiments were carried out under aerobic conditions at room temperatures. Five different water samples were used for the sorption experiments; two water samples from the tunnels with high DOM concentrations (U12n Tunnel and U12t Tunnel), two tunnel water samples with the DOM removed (U12n low DOM and U12t low DOM), and one groundwater with a naturally low DOM concentration (ER-12-3). All the water samples were allowed to equilibrate with the zeolitized tuff samples for one week prior to the addition of the radionuclides. The liquid/solid ratio used was 45 mL solution /0.3g solid for the Pu(IV) and Am(III) experiments and 45mL solution/3g solid for the Np(V) and U(VI) experiments. The resulting pH of water in each reactor after a week of equilibration with zeolitized tuff was used as a reference value to guide pH adjustment after addition of radionuclide stock solution. An appropriate amount of NaOH solution was added to each sorption tube prior to the introduction of the acidic radionuclide stock solution, so that the excess acid from the isotope stock solution was neutralize as soon as it was added into the sorption solution. The pH was adjusted to within 0.5 pH units of the reference value and was measured at each sampling date. No attempt was made to control effects of photo synthesis/catalysis and redox by the bacteria in waters for these batch sorption experiments. Two samples (at approximately one-day and 30 days) were collected for each batch experiment. During sampling, each sample was centrifuged at 4500 rpm for 10-15 minutes to separate the majority of the solids from the liquid. An aliquot of < 2 mL fluid was then

removed from the top of the 50mL centrifuge tube and placed into a microcentrifuge tube. The fluid was centrifuged at 10,000 rpm for 1 hour to remove any remaining colloids 30 nm or larger from the supernatant. An aliquot of the supernatant was taken from the top of the sample for analyses. A Tri-Carb 2600XL Liquid Scintillation Analyzer (Packard Instruments) was used for alpha liquid scintillation counting of the dissolved ^{241}Am, ^{238}Pu and ^{233}U in the supernatant. A quadrupole ICP-MS was used for analysis of dissolved ^{237}Np. Table 1 presents the initial conditions for each set of sorption experiments.

Table 1. Initial conditions used in the sorption experiments

Radionuclide	Oxidation State	Initial α-activity in fluid, dpm/g	Initial conc., M	Solid mass, g	Liquid mass, g
^{241}Am	III	815	4.5E-10	0.3	45
^{238}Pu	IV	830	1.0E-10	0.3	45
^{237}Np	V	1.1	3.0E-09	3	45
^{233}U	VI	127	2.6E-08	3	45

3. Results

The data are tabulated in appendix A. The perched water samples collected from U12n Tunnel and U12t Tunnel in 2006 are characterized by high carbonate concentration (55-80 mg carbon/L), mildly reducing conditions, and high dissolved organic carbon concentrations (>25 ppm C) (Stoller-Navarro Joint Venture, 2007). This unusual water chemistry can be attributed to anthropogenic sources of organic carbon as well as microbial activity in the perched tunnel water.

3.1 Characterization of volcanic zeolitized tuff core

The mineralogy and BET surface area of volcanic tuff rocks used in sorption experiments was determined. Table 2 lists information on the four volcanic zeolitized tuff samples. The mineralogy of all four zeolitized tuff samples are similar; the major phases in each rock sample are quartz, feldspar and zeolites. The mineralogy is typical of the zeolitized tuffs located at level in U12n Tunnel and U12t Tunnel.

The BET measurements in Table 2 show that volcanic zeolitized tuff sample UE12t #2 has the lowest surface area and UE12n #15A has the highest. The XRD results show that the primary components in all four zeolitized tuff samples are quartz, feldspars and zeolites, consistent with the lithologic description of these rocks. Quartz was present in all four samples, but sample UE12n #8 has much less than the other samples. Zeolites and feldspars were also observed in all four samples. The zeolites are likely to be clinoptilolite and /or heulandite according to the XRD pattern database, but no attempt was made to further identify the feldspars and zeolites.

Table 2. Volcanic zeolitized tuff core information

Hole	Interval, m	Geologic Unit*	Mineralogy	Surface Area ($m^2.g^{-1}$)
UE-12t #2	429.8-430.0	Tuff; ash-fall; pale greenish-yellow, pale to moderate red, & pale grayish-pink; zeolitized; silicified; fine to coarse grained; contains sparse to moderate, fine to coarse, lithic fragments & pumice. [Tn2, Indian Trail Fm.]	Quartz, feldspar, zeolites	5.1
UE-12t #4	390.8-390.9	Tuff; ash-fall, reworked ash-fall, peralkaline ash-fall, & tuffaceous sandstone; grayish-yellow, yellowish-gray, grayish-pink, moderate greenish-yellow; thin to thick bedded; zeolitized, several silicified zones, few argillized zones. [Tn 4CD,4AB]	Quartz, feldspar, zeolites	14.4
UE-12n #8	388.3-388.5	Tuff; alternately calc-alkaline reworked ash-fall, ash-fall, peralkaline ash-fall & reworked ash-fall, & tuffaceous sandstone; grayish-orange-pink, moderate red, & yellowish-gray; thin bedded; zeolitized; competent; contains several thin silicified beds. [Tn3 BC]	Feldspar, zeolites, quartz	19.2
UE-12n 15A	377.4-377.6	Tuff; calc-alkaline ash-fall, some peralkaline ash-fall, minor reworked ash-fall. [Tn 4J]	Quartz, feldspar, zeolites	34.9

* Magner (2007)

3.2 Characterization of water samples

Water samples were analyzed for major cations, anions, organic and inorganic carbon (Table 3). Groundwater collected from well ER-12-3, located 2100 m southwest of entrance to U12n Tunnel, has a DOM concentration nearly two orders of magnitude lower than water from U12n Tunnel and U12t Tunnel. The DOM level in the activated charcoal treated tunnel water samples is comparable to ER-12-3 water. However, the ER-12-3 water from a carbonate rock aquifer (LCA) has higher Ca and Mg and lower Na than the tunnel samples. The pH of all the water samples reported in Table 3 are similar, ranging from 8.2-.8.9.

Treating tunnel samples with activated charcoal to remove DOM resulted in higher sulfate concentrations. To test the effect of sulfate on the sorption of actinides to volcanic tuff, eight duplicate sorption experiments were carried out in low sulfate solutions. The results indicate that the sorption of Am(III) and Pu(IV) in waters with both high and low sulfate concentrations are the same within experimental errors (data are shown in Appendix A). Pu(IV) and Am(III) K_d values were not affected by the increased sulfate concentrations.

The DOM extracts of the water samples from the tunnels were analyzed by GC-MS. The GC-MS can only examine the volatile fraction of organic compounds with molecular weights of 450 or less. The chromatograms are shown in Figure 3. The volatile fraction (<450 molecular weight) comprised only a little more than 2% of the total dissolved organic matter in the tunnel waters. Further work is needed to characterize the rest of DOM in these tunnel waters. The data in Figure 3 suggest that these two tunnel water samples contain a similar family of low molecular weight organic compounds, which represent typical unresolved complex mixtures (UCM) of alkyl and aromatic hydrocarbons (Boot et al., 2007).

Table 3. Water analysis of samples from U12-t and U12n tunnels and water well ER-12-3

Water Sample	pH	Cl^- mg/L	SO_4^{2-} mg/L	Na mg/L	K mg/L	Mg mg/L	Ca mg/L	DIC* mg C/L	DOC** mg C/L
ER-12-3	8.29	6.1	29.0	32	2.0	10.1	14.9	23.1	0.2
U12n Main	8.76	14.6	32.5	143	4.2	< 1	2.5	55.6	14.9
U12n low DOM organics removed (1st batch)	8.28	25.7	143.4	151	0.3	< 1	< 0.1	30.4	0.3
U12n low DOM organics removed (2nd batch)	8.75	13.8	40.7	135	5.8	0.4	1.6	52.8	0.3
U12t Plug	8.85	21.7	107.2	236	6.7	< 1	3.2	79.0	18.5
U12t low DOM organics removed (1st batch)	8.26	27.6	211.2	239	3.0	< 1	< 0.1	56.6	0.4
U12t low DOM organics removed (2nd batch)	8.85	23.4	141.8	220	5.6	0.1	0.4	68.0	0.2

Notes: *dissolved inorganic carbon, ** dissolved organic carbon. The sulfate concentration increased because of the use of charcoal columns for removal of organics (1st batch) and was later decreased after treating charcoal columns with $NaHCO_3$ (2nd batch).

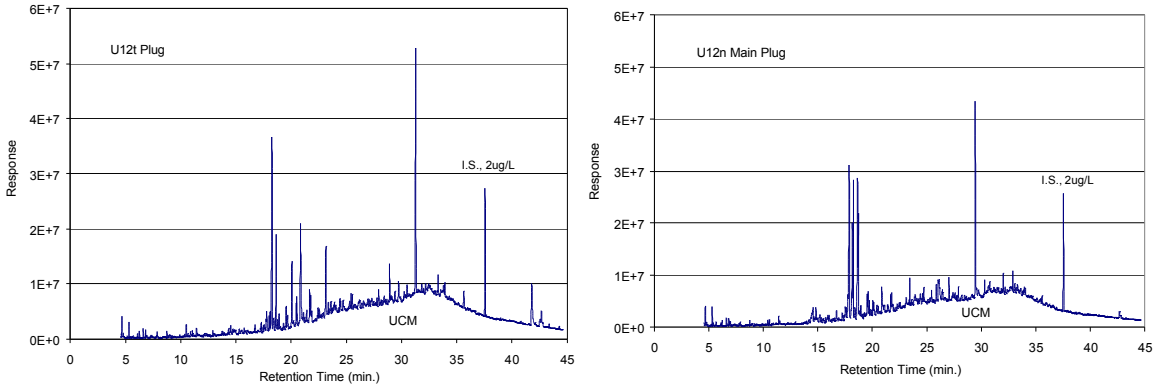

Figure 3. Total gas chromatograms extracts from U12t tunnel and U12n tunnel water. Booth et al. (2007) suggests that these complex mixtures most likely consist of alkyl and aromatic hydrocarbons.

The potential sorption of DOM to the volcanic zeolitized tuff samples was investigated by equilibrating three grams of zeolitized tuff with 45mL of waters for one month. The fluid was separated from the solid and analyzed for DIC and DOC. The results of the water analyses are listed in Table 4. The concentrations of DIC and DOC before and after the sorption experiments are similar, suggesting that there was little to no sorption of the DOM to the zeolitized tuff during the experiments. The stable DOM concentrations also

suggested that microbial/biological activity during the batch sorption experiments were not expected to have significant effects on the actinides sorption results.

Table 4. DIC and DOC content of water samples equilibrated with volcanic zeolitized tuff.

Tunnel water/rock	Supernatant separation methods	DIC	DOC	DIC	DOC
		Before Equilibration		After Equilibration	
		---------- mg C/L ----------			
U12t-plug/UE12t #2	Supernatant filtered through 20 nm pore size syringe filter	79	18.5	82	19.1
U12t-plug/UE12t #4				83	19.0
U12n Main/UE12n #8	centrifuge @4500 rpm for 140 min. particle size < 100 nm	56	14.9	59	15.0
U12n Main/UE12n 15A				55	13.1

3.3 Sorption of the radionuclides

3.3.1 Am(III) sorption

The sorption of Am(III) on four different rock samples (UE-12t#2, UE-12r#4, UE-12n#8, UE-12n#15A) was conducted over a period of 33 days. Three different waters for each zeolitized tuff sample were used: ER-12-3 with naturally low DOM, U12t tunnel or U12n tunnel with high DOM and U12t low DOM or U12n low DOM as DOM removed. The Am(III) K_d values for the four volcanic zeolitized tuff samples investigated are plotted on a log scale in Figure 4. Two aliquots from each sorption experiment were taken for analyses of dissolved Am(III). The first sample was collected after 3 days and the second after 33 days. The average pH of the solutions is also indicated on the plot. Appendix A tabulates the complete sampling data collected from Am sorption experiments.

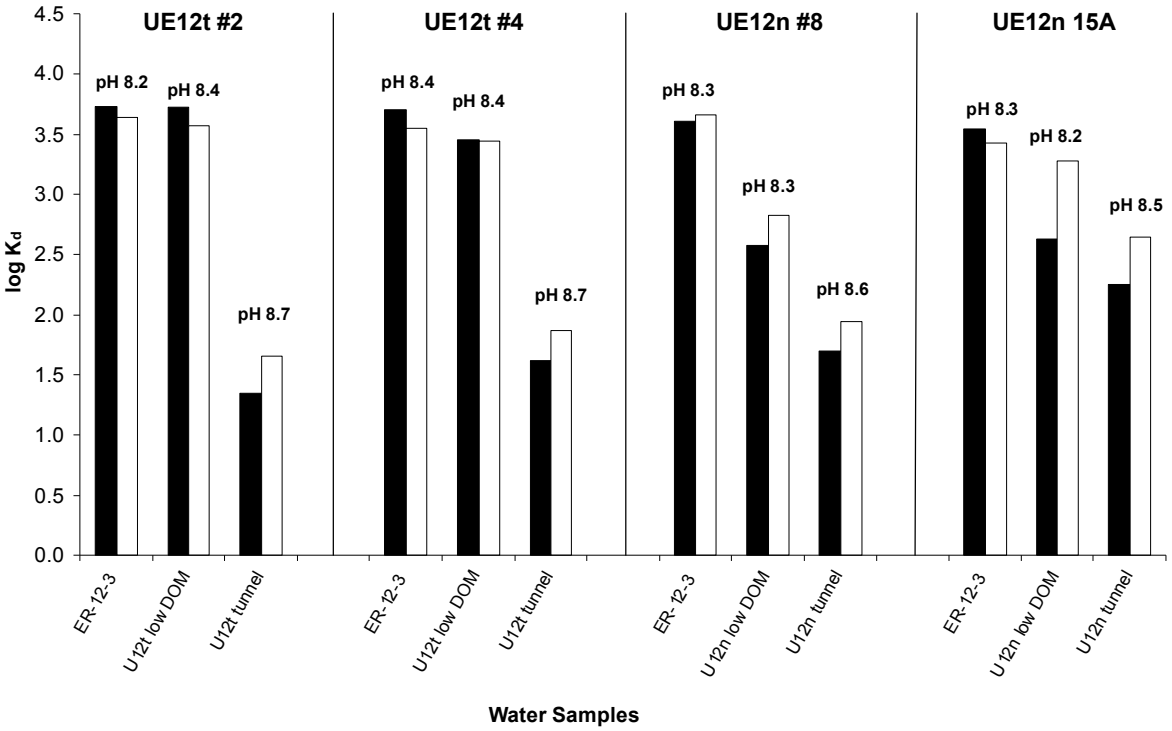

Figure 4. Comparison of Am(III) sorption in water from well ER-12-3, U12t tunnel and U12n tunnel with high and low DOM. ER-12-3 is a groundwater with naturally low DOM. K_d values collected after 2 days (■) and 33 days (□) for four zeolitized tuff samples (UE12t #2, UE12t #4, UE12n #8 and UE12n 15A).

The presence of a high concentration of DOM in the tunnel waters significantly decreases the Am(III) K_d, increasing the mobility of Am. The three waters that have low DOM concentrations (ER-12-3, U12t low DOM and U12n low DOM) result in much higher Am(III) K_d values. The K_d for Am(III) measured in U12t tunnel water (high DOM) with UE-12t #2 and UE-12t #4 zeolitized tuff is ~2 log units lower than in naturally low DOM water ER-12-3 or in tunnel waters that had the DOM removed. The decrease of K_d is somewhat smaller in U-12n water with UE-12n #8 and UE-12n 15A zeolitized tuff. Nevertheless, the K_d decrease resulting from the presence of high DOM concentration is quite dramatic. The sorption K_d values between the beginning and end of the experiments are similar, suggesting reaction kinetics were fast. In general, pH values decreased only slightly over the period of the sorption experiments; from 0.1 to 0.5 pH.

3.3.2 Pu(IV) sorption

Similar to the Am(III) experiments, the sorption of Pu(IV) was conducted over a period of 31 days. Two samples from each solution were taken and analyzed for soluble Pu; one sample collected after 6 days and another sample collected after 31 days. Figure 5 shows the log K_d of Pu for each of the four volcanic tuff samples. Again, three different waters for

each zeolitized tuff sample were used: ER-12-3 with naturally low DOM, U12t tunnel or U12n tunnel with high DOM, and U12t low DOM or U12n low DOM as DOM removed. The average pH of the solutions is also indicated on the plot. Appendix B tabulates the complete sampling data collected from Pu sorption experiments.

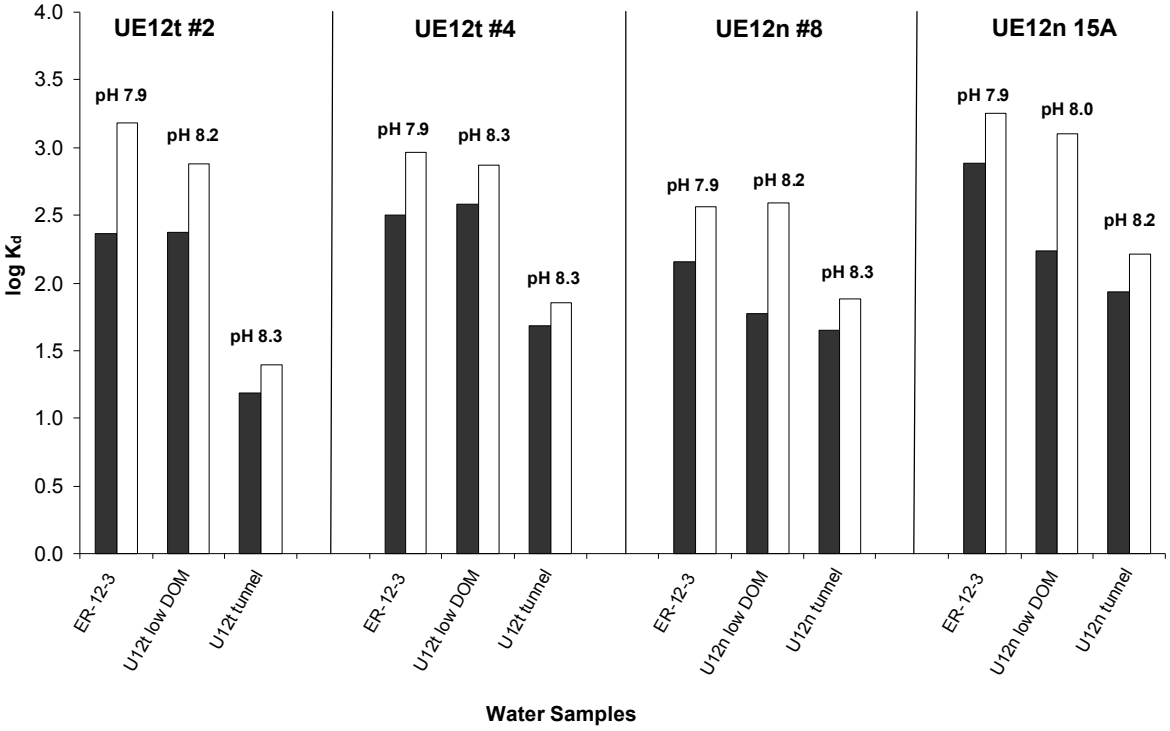

Figure 5. Comparison of Pu(IV) sorption in water from well ER-12-3, U12t tunnel and U12n tunnel with high DOM and low DOM. ER-12-3 is a groundwater with naturally low DOM. K_d values collected after 6 days (■) and 31 days (□) for four zeolitized tuff samples (UE12t #2, UE12t #4, UE12n #8 and UE12n 15A).

As seen in the Am(III) sorption experiments, the presence of high DOM concentration also decreases the K_d for Pu(IV) and increases the mobility of Pu in water. The three samples that have low concentrations of DOM (ER-12-3 and U12t low DOM and U12n low DOM) have higher K_d than the samples with high DOM. In the U-12t experiments, the K_d of Pu decreases approximately 1.0 to 2.0 log units. The sorption K_d values at the end of the experiment are higher than those measured in the beginning of the experiment, suggesting it may take longer time for Pu systems to reach equilibrium than that for Am system. In general, pH values decreased 0.3 pH unit over the period of the sorption experiment, which may have had some effect on the apparent amount of Pu sorption observed.

3.3.3 Np(V) sorption

The Np(V) sorption experiments were conducted over a period of 31 days. Figure 6 plots the log K_d of Np(V) for the four zeolitized tuff samples using the three different waters: ER-12-3 with naturally low DOM, U12t tunnel or U12n tunnel with high DOM and U12t low DOM or U12n low DOM as DOM removed. Samples were taken twice, once after 1 day and a second time after 31 days. The average pH of solutions is also shown on the plot. Appendix C tabulates the complete sampling data collected from Np sorption experiments.

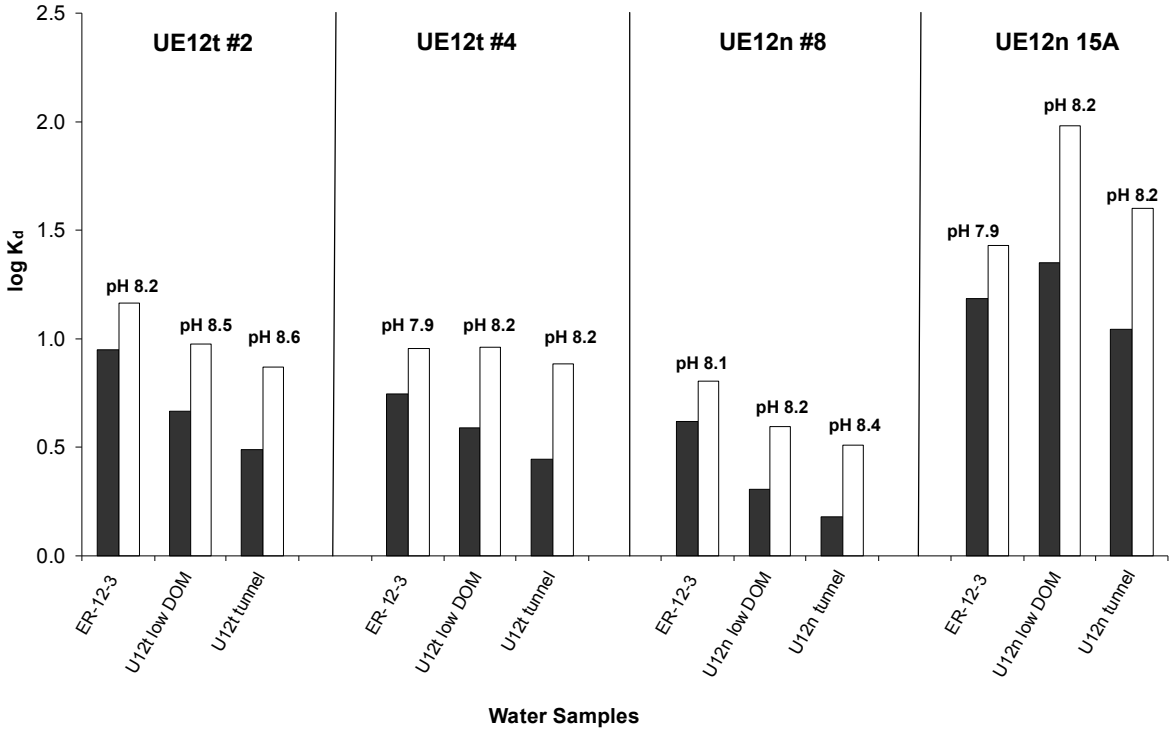

Figure 6. Comparison of Np(V) sorption in water from well ER-12-3, U12t tunnel and U12n tunnel with high DOM and low DOM. ER-12-3 is a groundwater with naturally low DOM. K_d values collected after 1 day (■) and 31 days (□) for four zeolitized tuff samples (UE12t #2, UE12t #4, UE12n #8 and UE12n 15A).

The presence of high concentrations of DOM decreases the Np(V) K_d in tunnel waters to a lesser extent than observed for Am(III) and Pu(IV). The K_d for Np(V) in water with a high DOM concentration decreases 0.3 and 0.1 log units on zeolitized tuff UE 12t #2 and UE 12t #4, respectively. In water with high DOM (U-12n tunnel), the K_d decreased approximately 0.1 and 0.4 log units with tuff samples UE12n #8 and UE12n 15A, respectively. The Np(V) K_d values increased over 31 days indicating that Np(V) sorption slowly reached its equilibrium. The pH of the Np(V) solutions decreased slightly over the

period of the experiment, between 0.0 to 0.4 pH units, which may have had some effect on the apparent sorption amount observed.

3.3.4 U(VI) sorption

The U(VI) sorption experiments were conducted over a period of 31 days. Figure 7 shows log K_d of U(VI) on four different volcanic zeolitized tuff samples using three different waters: ER-12-3 with naturally low DOM, U12t tunnel or U12n tunnel with high DOM and U12t low DOM or U12n low DOM as DOM removed. Samples were taken after one day and 31 days. The average pH of sorption solutions is also indicated on the plot. Appendix D tabulates the complete sampling data from U sorption experiments.

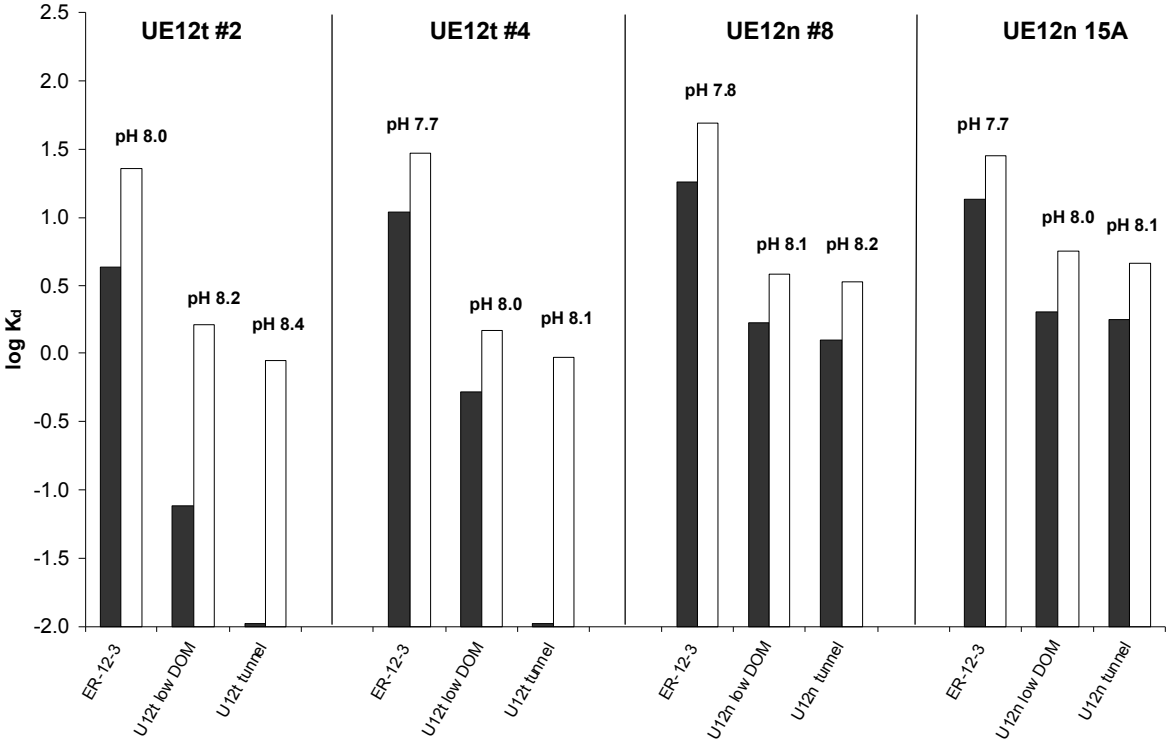

Figure 7. Comparison of U(VI) sorption in water from well ER-12-3, U12t tunnel and U12n tunnel with high and low DOM. ER-12-3 is a groundwater with naturally low DOM. K_d values collected after 1 day (■) and 31 days (□) for four zeolitized tuff samples (UE12t #2, UE12t #4, UE12n #8 and UE12n 15A).

Very weak U(VI) sorption was observed in ER-12-3 water. Even weaker or essentially no U(VI) sorption was observed in the water samples with high DOM concentrations. The observed K_d values in tunnel waters with high and low DOM are similar for all four zeolitized tuff samples after 31 days. The dissolved organic carbon may play a less important role for U(VI) sorption, compared to Am(III), Pu(IV) and Np(V). Although pH values of the solutions decreased slightly over time, the changes of pH during the experiment were minimal (0-0.2 pH unit). The results suggest that differences in water

chemistry between ER-12-3 and tunnel waters other than DOM concentration play a prominent role in controlling U(VI) K_d values. Our calculations suggested that the formation of soluble carbonate complexes would be >30% more in water U12t tunnel than those in groundwater ER-12-3. The K_d values obtained for U decreased as DIC increased in waters U12t tunnel, U12n and ER-12-3, respectively. Brady et al. (1999) reported that U-carbonate complexation leads to desorption of uranium from mineral surface, while Am-carbonate and maybe Pu-carbonate complexes appeared to sorb onto the surface. Our results in the report were consistent with their findings. For U(VI), it is likely that the high *dissolved inorganic carbon* (DIC) plays a more important role in controlling K_d than the DOC (Zavarin and Bruton 2004a; 2004b).

5. Concluding Remarks

Am and Pu sorption to volcanic zeolitized tuff is strongly dependent on the dissolved organic matter (DOM) concentration in groundwater. K_d values of strongly sorbing species, such as Am (III) and Pu(IV),decrease by up to two orders of magnitude in the presence of DOM. In contrast, sorption of Np(V) is less affected by DOM. Only a slight decrease in K_d was observed for Np(V) sorption in water with a high DOM compared to low DOM. Little to no DOM effects on U(VI) sorption were observed. U(VI) sorption is very strongly dependent on the dissolved inorganic carbon concentration in groundwater. Based on a comparison of ER-12-3 and tunnel water, it is apparent that the high DIC in the tunnel waters reduces U(VI) K_d values more effectively than the DOC.

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Appendix A: Am Sorption Data Table

		Date	3/12/2007	3/12/2007	3/19/2007	3/21/2007			3/23/2007	3/23/2007	3/23/2007	3/23/2007	4/23/2007	4/23/2007	4/23/2007	4/23/2007	4/23/2007
S#	RN	Sample ID	solid mass, g	Water mass, g	pH before Am spike	spike 241Am, g	initial dpm/g	total volume prior to 1st sampling, mL	pH @ 1st sampling	1st sampling dpm/g	**%sorbed Am @ 1st sampling**	**log Kd**	total volume prior to 2nd sampling	pH @ 2nd sampling	2nd sampling dpm/g	**%sorbed Pu @ 2nd sampling**	**log Kd**
									Average,								
0	^{241}Am	^{241}Am in 2% HNO3 solution		45.70		0.0812	815.5		dpm/g	813.5							
		^{241}Am in 2% HNO3 solution							815.5	817.6				blank			
		^{241}Am in 2% HNO3 solution centrifuged								823.2					822.3		
		^{241}Am in 2% HNO3 solution centrifuged							814.5	805.7					823.6		
1	^{241}Am	ER-12-3 Blank		45.23	8.23	0.081	821.9	45.40	8.24	300.3	**63.5%**		42.70	8.13	208.0	**74.7%**	
4	^{241}Am	ER-12-3 UE12t #2-1	0.301	45.12	8.19	0.0825	839.1	45.29	8.29	21.1	**97.5%**	**3.8**	42.59	8.04	25.7	**96.9%**	**3.7**
5	^{241}Am	ER-12-3 UE12t #2-2	0.3056	45.10	8.18	0.0814	828.4	45.27	8.29	23.6	**97.2%**	**3.7**	43.57	8.05	26.5	**96.8%**	**3.6**
10	^{241}Am	ER-12-3 UE12t #4-1	0.3009	45.08	8.07	0.0821	835.7	45.26	8.44	23.2	**97.2%**	**3.7**	42.56	8.16	31.9	**96.2%**	**3.6**
11	^{241}Am	ER-12-3 UE12t #4-2	0.3069	45.07	8.10	0.0814	828.8	45.24	8.69	24.5	**97.0%**	**3.7**	43.54	8.18	31.2	**96.2%**	**3.6**
24	^{241}Am	ER-12-3-UE12n #8-1	0.3041	45.02	8.30	0.0813	828.8	45.19	8.4	27.4	**96.7%**	**3.6**	42.49	8.03	25.3	**96.9%**	**3.6**
25	^{241}Am	ER-12-3-UE12n #8-2	0.3048	44.98	8.26	0.0822	838.7	45.15	8.46	31.8	**96.2%**	**3.6**	43.45	8.11	24.5	**97.1%**	**3.7**
16	^{241}Am	ER-12-3-UE12n 15A-1	0.3072	45.05	8.21	0.0818	833.4	45.22	8.33	32.7	**96.1%**	**3.6**	42.52	8.14	41.8	**95.0%**	**3.4**
17	^{241}Am	ER-12-3-UE12n 15A-2	0.3066	45.05	8.16	0.0823	838.4	45.22	8.42	34.9	**95.8%**	**3.5**	43.52	8.18	41.9	**95.0%**	**3.4**
2	^{241}Am	T-Plug Blank		45.13	8.63	0.081	823.7	45.30	8.69	821.1	**0.3%**		43.60	8.56	847.0	**-2.8%**	
6	^{241}Am	T-Plug-UE12t #2-1	0.3044	44.94	8.67	0.0801	818.0	45.11	8.79	717.6	**12.3%**	**1.3**	43.41	8.52	623.0	**23.8%**	**1.6**
7	^{241}Am	T-Plug-UE12t #2-2	0.3014	44.91	8.70	0.0805	822.6	45.08	8.73	711.2	**13.5%**	**1.4**	43.38	8.56	623.9	**24.2%**	**1.7**
12	^{241}Am	T-Plug-UE12t #4-1	0.3006	44.87	8.69	0.0803	821.4	45.04	8.74	640.3	**22.0%**	**1.6**	43.34	8.6	547.7	**33.3%**	**1.9**
13	^{241}Am	T-Plug-UE12t #4-2	0.3048	44.95	8.66	0.0805	821.9	45.12	8.74	642.4	**21.8%**	**1.6**	43.42	8.56	536.5	**34.7%**	**1.9**
3	^{241}Am	T-Plug ACT1 Blank		45.09	8.33	0.0815	829.5	45.26	8.38	733.4	**11.6%**		43.56	8.3	638.1	**23.1%**	
8	^{241}Am	T-Plu ACT1-UE12t #2-1	0.3052	45.01	8.31	0.0814	829.9	45.18	8.49	24.3	**97.1%**	**3.7**	43.48	8.35	30.7	**96.3%**	**3.6**
9	^{241}Am	T-Plu ACT1-UE12t #2-2	0.3002	44.99	8.30	0.0802	818.1	45.16	8.55	21.4	**97.4%**	**3.7**	43.46	8.37	31.1	**96.2%**	**3.6**
14	^{241}Am	T-Plu ACT1-UE12t #4-1	0.3057	45.00	8.39	0.0819	835.2	45.17	8.57	41.4	**95.0%**	**3.5**	43.47	8.32	40.0	**95.2%**	**3.5**
15	^{241}Am	T-Plu ACT1-UE12t #4-2	0.3041	44.95	8.41	0.0815	832.0	45.13	8.47	42.0	**95.0%**	**3.4**	43.43	8.29	41.1	**95.1%**	**3.4**
18	^{241}Am	N-Main Blank		44.98	8.73	0.0813	829.5	45.15	8.77	816.6	**1.6%**		43.45	8.39	820.1	**1.1%**	
20	^{241}Am	N-Main-UE12n 15A-1	0.3046	44.94	8.64	0.0804	821.0	45.11	8.63	371.5	**54.8%**	**2.3**	43.41	8.37	199.8	**75.7%**	**2.6**
21	^{241}Am	N-Main-UE12n 15A-2	0.3069	44.92	8.63	0.0814	831.6	45.10	8.72	377.9	**54.5%**	**2.2**	43.40	8.47	200.6	**75.9%**	**2.6**
26	^{241}Am	N-Main-UE12n #8-1	0.3006	44.87	8.65	0.0808	826.5	45.04	8.67	614.1	**25.7%**	**1.7**	43.34	8.48	503.0	**39.1%**	**2.0**
27	^{241}Am	N-Main-UE12n #8-2	0.3018	45.39	8.66	0.0808	817.0	45.56	8.67	618.2	**24.3%**	**1.7**	43.86	8.46	516.6	**36.8%**	**1.9**
19	^{241}Am	N-Main ACT1 Blank		45.02	8.18	0.0809	824.7	45.19	8.18	834.4	**-1.2%**		43.49	8.11	645.0	**21.8%**	
22	^{241}Am	N-Main ACT1-UE12n 15A-1	0.3034	44.94	8.08	0.0813	830.3	45.11	8.29	222.1	**73.3%**	**2.6**	43.41	8.16	57.8	**93.0%**	**3.3**
23	^{241}Am	N-Main ACT1-UE12n 15A-2	0.3032	44.94	8.09	0.0818	835.4	45.11	8.31	208.2	**75.1%**	**2.7**	43.41	8.17	57.5	**93.1%**	**3.3**
28	^{241}Am	N-Main ACT1-UE12n #8-1	0.3031	44.93	8.17	0.0812	829.4	45.10	8.38	228.4	**72.5%**	**2.6**	43.40	8.13	146.5	**82.3%**	**2.8**
29	^{241}Am	N-Main ACT1-UE12n #8-2	0.3028	44.90	8.15	0.0807	824.8	45.07	8.34	240.1	**70.9%**	**2.6**	43.37	8.15	144.9	**82.4%**	**2.8**
			4/17/2007	4/17/2007		4/25/2007	4/25/2007	4/27/2007	4/27/2007	4/27/2007	4/27/2007	4/27/2007	5/30/2007	5/30/2007	5/30/2007	5/30/2007	5/30/2007
LS#14	^{241}Am	T-Plu ACT2-UE12t #4-1	0.3074	44.93		0.0818	835.6	45.10	8.37	70.7	**91.5%**	**3.2**	43.30	8.28	35.66	**95.7%**	**3.5**
LS#15	^{241}Am	T-Plu ACT2-UE12t #4-2	0.301	44.90		0.0816	834.1	45.07	8.39	55.4	**93.4%**	**3.3**	43.27	8.29	26.70	**96.8%**	**3.6**
LS#28	^{241}Am	N-Main ACT2-2-UE12n #8-1	0.3027	44.92		0.0813	830.6	45.09	8.36	137.0	**83.5%**	**2.9**	43.29	8.23	65.98	**92.1%**	**3.2**
LS#29	^{241}Am	N-Main ACT2-2-UE12n #8-2	0.303	44.94		0.0808	825.1	45.11	8.32	141.3	**82.9%**	**2.9**	43.31	8.16	56.29	**93.2%**	**3.3**

Sample list, 45 mL of water and 0.3g gram of solid per sample

1st sampling: 3/23/07 (one day sorption) Centrifuge samples in 45mL test tube @ 5200 rpm for 20 min. Transfer 1.7 mL supernatant into 2mL microcentrifuge tube and spin at 10,000 rpm for 100 min. Take 1.5mL supernatant for LSC.

2nd sampling: 4/23/07 (one month sorption) Centrifuge samples in 45mL tet tube @3420 rpm for 10 min. Take 1.8mL supernatant into 2mL microcentrifuge tube and centrifuge at 10, 000 rpm for 100min. Take 1.5mL supernatant for LSC. Particle size cutoff was 21 nm.

Appendix B: Pu Sorption Data Table

		Date	3/8/2007	3/8/2007	3/14/2007	3/19/2007		3/20/2007	3/21/2007	3/20/2007	3/20/2007	3/20/2007	3/25/2007	3/25-26/07	3/25/2007	3/25/2007	3/25/2007	4/19/2007	4/19/2007	4/19/2007	4/19/2007	4/19/2007
S#	RN	Sample ID	solid mass, g	Water mass, g	pH before Pu spike	spike ^{238}Pu, g	initial dpm/g	1st sampling mL	pH @ 1st sampling	1st sampling dpm/g	**%sorbed Pu @1st sampling**	**log Kd**	2nd sampling, mL	pH @ 2nd sampling	2nd sampling dpm/g	**%sorbed Pu @ 2nd sampling**	**log Kd**	3rd sampling, mL	pH @ 3rd sampling	3rd sampling dpm/g	**%sorbed Pu @ 3rd sampling**	**log Kd**
0	^{238}Pu	^{238}Pu in 2% HNO3 solution		45.7035		0.1018	830.9							**Average**	827.4							
		^{238}Pu in 2% HNO3 solution												828.8	830.3							
		^{238}Pu in 2% HNO3 solution centrifuged													819.7			1.5		809.8		
		^{238}Pu in 2% HNO3 solution centrifuged												820.2	820.7			1.5		816.2		
1	^{238}Pu	ER-12-3 Blank		44.9703	8.08	0.1008	834.1	3.00	7.04	650.6	**22.0%**		1.5	**7.96**	551.0	**33.9%**		1.5	**7.95**	498.8	40.2%	
4	^{238}Pu	ER-12-3 UE12t #2-1	0.3014	44.9100	8.25	0.1014	840.1	3.00	7.06	498.3	**40.7%**	**2.0**	1.5	**8.01**	274.4	**67.3%**	**2.5**	1.5	**7.99**	55.2	93.4%	**3.3**
5	^{238}Pu	ER-12-3 UE12t #2-2	0.3048	45.0188	8.16	0.0989	817.5	3.00	6.98	541.9	**33.7%**	**1.9**	1.5	**7.89**	345.4	**57.7%**	**2.3**	1.5	**7.89**	80.2	90.2%	**3.1**
10	^{238}Pu	ER-12-3 UE12t #4-1	0.3116	45.0312	8.34	0.1016	839.5	3.00	7.07	404.1	**51.9%**	**2.2**	1.5	**8.00**	252.1	**70.0%**	**2.5**	1.5	**7.83**	104.1	87.6%	**3.0**
11	^{238}Pu	ER-12-3 UE12t #4-2	0.3024	45.0916	8.14	0.1013	836.0	3.00	7.13	416.8	**50.1%**	**2.2**	1.5	**7.86**	257.6	**69.2%**	**2.5**	1.5	**7.94**	104.4	87.5%	**3.0**
18	^{238}Pu	ER-12-3-UE12n 15A-1	0.3082	45.0661	8.07	0.1041	859.5	3.00	7.08	246.9	**71.3%**	**2.6**	1.5	**7.92**	128.3	**85.1%**	**2.9**	1.5	**7.88**	61.9	92.8%	**3.2**
19	^{238}Pu	ER-12-3-UE12n 15A-2	0.2998	45.0743	8.12	0.1002	827.2	3.00	7.09	284.9	**65.6%**	**2.5**	1.5	**8.00**	131.0	**84.2%**	**2.9**	1.5	**7.95**	55.2	93.3%	**3.3**
24	^{238}Pu	ER-12-3-UE12n #8-1	0.3008	45.0511	8.22	0.0978	807.9	3.00	7.13	496.7	**38.5%**	**2.0**	1.5	**8.08**	395.0	**51.1%**	**2.2**	1.5	**7.93**	203.3	74.8%	**2.6**
25	^{238}Pu	ER-12-3-UE12n #8-2	0.3044	45.0600	8.34	0.099	817.6	3.00	7.03	490.7	**40.0%**	**2.0**	1.5	**7.95**	409.4	**49.9%**	**2.1**	1.5	**7.83**	228.1	72.1%	**2.5**
2	^{238}Pu	T-Plug Blank		45.0480	8.48	0.0999	825.2	3.00	7.8	814.8	**1.3%**		1.5	**8.41**	827.8	**-0.3%**		1.5	**8.32**	810.6	1.8%	
6	^{238}Pu	T-Plug-UE12t #2-1	0.2975	45.0499	8.62	0.0994	821.1	3.00	7.74	776.6	**5.4%**	**0.9**	1.5	**8.38**	757.0	**7.8%**	**1.1**	1.5	**8.19**	706.1	14.0%	**1.3**
7	^{238}Pu	T-Plug-UE12t #2-2	0.3019	45.0256	8.66	0.1044	862.7	3.00	7.78	782.6	**9.3%**	**1.2**	1.5	**8.47**	758.0	**12.1%**	**1.3**	1.5	**8.32**	716.0	17.0%	**1.4**
12	^{238}Pu	T-Plug-UE12t #4-1	0.3066	45.0649	8.53	0.1012	835.6	3.00	7.98	708.4	**15.2%**	**1.4**	1.5	**8.51**	604.4	**27.7%**	**1.7**	1.5	**8.3**	530.6	36.5%	**1.9**
13	^{238}Pu	T-Plug-UE12t #4-2	0.3023	45.0514	8.56	0.1017	840.0	3.00	7.75	723.0	**13.9%**	**1.4**	1.5	**8.27**	636.8	**24.2%**	**1.6**	1.5	**8.24**	559.6	33.4%	**1.8**
3	^{238}Pu	T-Plug ACT1 Blank		45.1244	8.34	0.1001	825.5	3.00	7.56	738.7	**10.5%**		1.5	**8.40**	608.4	**26.3%**		1.5	**8.37**	299.6	63.7%	
8	^{238}Pu	T-Plu ACT1-UE12t #2-1	0.3031	45.0904	8.24	0.1008	831.9	3.00	7.49	431.2	**48.2%**	**2.1**	1.5	**8.25**	302.8	**63.6%**	**2.4**	1.5	**8.11**	117.6	85.9%	**2.9**
9	^{238}Pu	T-Plu ACT1-UE12t #2-2	0.3045	45.0932	8.20	0.101	833.5	3.00	7.47	459.9	**44.8%**	**2.1**	1.5	**8.35**	313.0	**62.4%**	**2.4**	1.5	**8.17**	133.5	84.0%	**2.8**
14	^{238}Pu	T-Plu ACT1-UE12t #4-1	0.2997	45.0532	8.27	0.1008	832.5	3.00	7.51	380.7	**54.3%**	**2.3**	1.5	**8.38**	242.8	**70.8%**	**2.5**	1.5	**8.17**	143.8	82.7%	**2.8**
15	^{238}Pu	T-Plu ACT1-UE12t #4-2	0.3021	45.0789	8.22	0.1023	844.4	3.00	7.46	351.6	**58.4%**	**2.3**	1.5	**8.36**	208.2	**75.3%**	**2.6**	1.5	**8.15**	113.5	86.6%	**2.9**
16	^{238}Pu	N-Main Blank		45.0261	8.63	0.1004	829.7	3.00	7.54	805.5	**2.9%**		1.5	**8.19**	821.3	**1.0%**		1.5	**8.19**	813.7	1.9%	
20	^{238}Pu	N-Main-UE12n 15A-1	0.3029	45.0352	8.58	0.1016	839.5	3.00	7.55	677.7	**19.3%**	**1.6**	1.5	**8.26**	513.7	**38.8%**	**1.9**	1.5	**8.14**	373.1	55.6%	**2.2**
21	^{238}Pu	N-Main-UE12n 15A-2	0.3006	45.0509	8.57	0.1007	831.8	3.00	7.58	684.9	**17.7%**	**1.5**	1.5	**8.36**	524.2	**37.0%**	**1.9**	1.5	**8.14**	380.0	54.3%	**2.2**
26	^{238}Pu	N-Main-UE12n #8-1	0.3038	45.0447	8.74	0.0998	824.5	3.00	7.55	727.8	**11.7%**	**1.3**	1.5	**8.46**	628.4	**23.8%**	**1.6**	1.5	**8.2**	530.8	35.6%	**1.9**
27	^{238}Pu	N-Main-UE12n #8-2	0.2998	45.0316	8.67	0.1002	828.0	3.00	7.56	725.3	**12.4%**	**1.3**	1.5	**8.35**	622.1	**24.9%**	**1.7**	1.5	**8.18**	517.7	37.5%	**1.9**
17	^{238}Pu	N-Main ACT1 Blank		45.0489	8.30	0.1005	830.1	3.00	7.05	814.5	**1.9%**		1.5	**8.04**	756.2	**8.9%**		1.5	**8.05**	634.4	23.6%	
22	^{238}Pu	N-Main ACT1-UE12n 15A-1	0.3046	45.0662	8.07	0.1003	828.2	3.00	7.07	606.1	**26.8%**	**1.7**	1.5	**8.08**	372.9	**55.0%**	**2.2**	1.5	**7.95**	74.2	91.0%	**3.1**
23	^{238}Pu	N-Main ACT1-UE12n 15A-2	0.3017	45.0481	8.12	0.1009	833.5	3.00	7.08	605.1	**27.4%**	**1.8**	1.5	**8.25**	368.8	**55.8%**	**2.2**	1.5	**7.87**	84.3	89.9%	**3.1**
28	^{238}Pu	N-Main ACT1-UE12n #8-1	0.33066	45.0568	8.40	0.102	842.4	3.00	7.29	716.8	**14.9%**	**1.4**	1.5	**8.45**	538.9	**36.0%**	**1.9**	1.5	**8.05**	172.9	79.5%	**2.7**
29	^{238}Pu	N-Main ACT1-UE12n #8-2	0.2992	45.0418	8.32	0.1019	841.8	3.00	7.14	738.7	**12.2%**	**1.3**	1.5	**8.23**	625.1	**25.7%**	**1.7**	1.5	**8.02**	251.5	70.1%	**2.5**
			4/17/2007	4/17/2007		4/25/2007	4/25/2007	4/27/2007	4/27/2007	4/27/2007	4/27/2007	4/27/2007	5/2/2007	5/2/2007				5/30/2007	5/30/2007			
LS#8	^{238}Pu	T-Plu ACT2-UE12t #2-1	0.30559	45.2552		0.1029	845.7	1.50	8.28	565.8	**33.1%**	**1.9**	1.5	8.30	321.7	**62.0%**	**2.4**	1.5	8.39	66.5	92.1%	**3.2**
LS#9	^{238}Pu	T-Plu ACT2-UE12t #2-2	0.3082	44.9495		0.1023	846.4	1.50	8.38	682.7	**19.3%**	**1.5**	1.5	8.37	376.4	**55.5%**	**2.2**	1.5	8.38	91.3	89.2%	**3.0**
LS#28	^{238}Pu	N-Main ACT2-2-UE12n #8-1	0.2982	45.0488		0.1028	848.7	1.50	8.18	620.3	**26.9%**	**1.7**	1.5	8.14	376.6	**55.6%**	**2.3**	1.5	8.21	75.4	91.1%	**3.2**
LS#29	^{238}Pu	N-Main ACT2-2-UE12n #8-2	0.308	44.9435		0.1023	846.5	1.50	8.19	618.7	**26.9%**	**1.7**	1.5	8.18	383.2	**54.7%**	**2.2**	1.5	8.16	74.6	91.2%	**3.1**

Sample list, 45 mL of water and 0.3g gram of solid per sample
1st sampling: 3/20/07 (one day sorption) Centrifuge samples in 45mL test tube @ 5200 for 30 - 60 min. Take 3mL of each supernatant for LSC.
2nd sampling: 3/25/07 (4 days after pH adjustment) centrifuge 45mL tubes @5200 rpm for 20 min. and transfer 1.8mL into a microcentrifuge tube and centrifuge @10,000rpm for 100 min. Pipette 1.5mL of the supernatant from LSC.
3rd sampling: 4/19/07 (one month sorptiont) centrifuge 45mL tubes @3420 rpm for 10 min. and transfer 1.8mL into a microcentrifuge tube and centrifuge @10,000rpm for 1000 min. Pipette 1.5mL of the supernatant from LSC.

Appendix C: Np Sorption Data Table

		Date	4/20/2007	4/20/2007	4/27/2007	4/27/2007	4/27/2007	4/28/2007	4/28/2007	4/28/2007	4/28/2007	4/28/2007	5/29/2007	5/29/2007	5/29/2007	5/29/2007	5/29/2007
S#	RN	Sample ID	solid mass g	Water mass g	pH before Np spike	spike ^{237}Np g	Initial Np conc., ppt	total volume prior to 1st sampling	pH @ 1st sampling	1st sampling ICP results ppt	**%sorbed Np @ 1st sampling**	**log Kd**	total volume prior to 2nd sampling	pH @ 2nd sampling	2nd sampling ICP results ppt	**%sorbed Np @ 2nd sampling**	**log Kd**
0	^{237}Np	^{237}Np in 2% HNO3 solution		45.3138		0.1019	714.0		Average, ppt	713.0							
		^{237}Np in 2% HNO3 solution							714.0	713.0							
		^{237}Np in 2% HNO3 solution centrifuged								714.0					705.5		
		^{237}Np in 2% HNO3 solution centrifuged								716.0					702.5		
1	^{237}Np	ER-12-3 Blank		45.0559	8.09	0.0976	687.8	45.17	8.06	689	**0.0%**		43.77	8.01	679	**1.28%**	
4	^{237}Np	ER-12-3 UE12t #2-1	3.0359	45.1554	8.47	0.0983	691.2	45.27	8.28	435	**37.1%**	**0.94**	43.87	7.98	355	**48.64%**	**1.15**
5	^{237}Np	ER-12-3 UE12t #2-2	3.0316	44.9444	7.98	0.1008	712.1	45.06	8.31	441	**38.1%**	**0.96**	43.66	8.03	352	**50.57%**	**1.18**
10	^{237}Np	ER-12-3 UE12t #4-1	3.0258	45.0665	7.85	0.1014	714.4	45.18	8.01	521	**27.1%**	**0.74**	43.78	7.73	445	**37.71%**	**0.96**
11	^{237}Np	ER-12-3 UE12t #4-2	3.0241	45.0125	7.56	0.1007	710.3	45.13	8	518	**27.1%**	**0.74**	43.73	7.8	443	**37.63%**	**0.95**
16	^{237}Np	ER-12-3-UE12n 15A-1	3.0241	45.0024	8.1	0.1033	728.8	45.12	7.97	360	**50.6%**	**1.18**	43.72	7.79	255	**65.04%**	**1.44**
17	^{237}Np	ER-12-3-UE12n 15A-2	3.029	44.9919	7.9	0.1025	723.3	45.11	8.02	355	**50.9%**	**1.19**	43.71	7.8	261	**63.92%**	**1.42**
24	^{237}Np	ER-12-3-UE12n #8-1	3.0299	45.1094	7.78	0.1023	720.0	45.23	8.28	565	**21.5%**	**0.61**	43.83	7.91	508	**29.45%**	**0.79**
25	^{237}Np	ER-12-3-UE12n #8-2	3.0214	45.2836	7.75	0.1021	715.9	45.40	8.28	557	**22.2%**	**0.63**	44.00	7.89	498	**30.43%**	**0.82**
2	^{237}Np	T-Plug Blank		45.0333	8.71	0.1012	713.5	45.15	8.78	720	**0.0%**		43.75	8.66	701	**1.75%**	
6	^{237}Np	T-Plug-UE12t #2-1	3.0142	44.8881	8.41	0.1019	720.7	45.01	8.68	594	**17.6%**	**0.50**	43.61	8.56	481	**33.25%**	**0.87**
7	^{237}Np	T-Plug-UE12t #2-2	3.0282	45.0062	8.53	0.1013	714.6	45.12	8.69	595	**16.7%**	**0.48**	43.72	8.57	478	**33.11%**	**0.87**
12	^{237}Np	T-Plug-UE12t #4-1	3.0015	45.2878	8.25	0.1016	712.3	45.41	8.28	601	**15.6%**	**0.45**	44.01	8.1	474	**33.46%**	**0.88**
13	^{237}Np	T-Plug-UE12t #4-2	3.0329	45.1283	8.25	0.1015	714.1	45.25	8.27	602	**15.7%**	**0.44**	43.85	8.1	470	**34.18%**	**0.89**
3	^{237}Np	T-Plug ACT2 Blank		44.9628	8.4	0.1023	722.4	45.08	8.63	718	**0.6%**		43.68	8.62	698	**3.37%**	
8	^{237}Np	T-Plu ACT2-UE12t #2-1	3.0144	44.96	8.55	0.1019	719.6	45.08	8.54	552	**23.3%**	**0.66**	43.68	8.5	441	**38.72%**	**0.98**
9	^{237}Np	T-Plu ACT2-UE12t #2-2	3.013	44.8384	8.4	0.1021	723.0	44.96	8.54	550	**23.9%**	**0.67**	43.56	8.4	441	**39.00%**	**0.98**
14	^{237}Np	T-Plu ACT2-UE12t #4-1	3.0321	44.9846	8.15	0.1021	720.6	45.10	8.22	571	**20.8%**	**0.59**	43.70	8.12	442	**38.66%**	**0.97**
15	^{237}Np	T-Plu ACT2-UE12t #4-2	3.0233	45.023	8.05	0.102	719.3	45.14	8.22	571	**20.6%**	**0.59**	43.74	8.04	453	**37.02%**	**0.94**
18	^{237}Np	N-Main Blank		45.0059	8.62	0.1013	714.6	45.12	8.74	719	**0.0%**		43.72	8.63	708	**0.93%**	
20	^{237}Np	N-Main-UE12n 15A-1	3.0259	44.9583	8.1	0.1018	718.9	45.08	8.22	414	**42.5%**	**1.04**	43.68	8.16	195	**72.82%**	**1.60**
21	^{237}Np	N-Main-UE12n 15A-2	3.0188	45.0726	8.12	0.1016	715.7	45.19	8.24	410	**42.7%**	**1.05**	43.79	8.17	196	**72.61%**	**1.60**
26	^{237}Np	N-Main-UE12n #8-1	3.0195	45.0963	8.35	0.1015	714.6	45.21	8.45	652	**8.8%**	**0.16**	43.81	8.29	591	**17.30%**	**0.50**
27	^{237}Np	N-Main-UE12n #8-2	3.025	45.1526	8.35	0.102	717.2	45.27	8.44	648	**9.7%**	**0.20**	43.87	8.27	586	**18.30%**	**0.53**
19	^{237}Np	N-Main ACT2-2 Blank		45.2087	8.25	0.1017	714.3	45.33	8.63	718	**0.0%**		43.93	8.59	702	**1.72%**	
22	^{237}Np	N-Main ACT2-2-UE12n 15A-1	3.025	44.9642	7.9	0.1019	719.5	45.08	8.2	309	**57.1%**	**1.30**	43.68	8.21	98	**86.37%**	**1.98**
23	^{237}Np	N-Main ACT2-2-UE12n 15A-2	3.0268	44.9446	7.93	0.1013	715.6	45.06	8.19	264	**63.1%**	**1.41**	43.66	8.2	96	**86.58%**	**1.98**
28	^{237}Np	N-Main ACT2-2-UE12n #8-1	3.0342	45.0227	8.24	0.1013	714.4	45.14	8.02	1252	**12.4%**	**0.32**	43.74	8.27	1144	**19.93%**	**0.57**
29	^{237}Np	N-Main ACT2-2-UE12n #8-2	3.0143	45.0525	8.25	0.1019	718.1	45.17	8.41	635	**11.6%**	**0.29**	43.77	8.28	562	**21.74%**	**0.62**

Sample list, 45 mL of water and 3g gram of solid per sample

1st sampling: 4/29/07 (two-day sorption) Centrifuge samples in 45mL test tube @ 3420 rpm for 10 min. Transfer 1.4 mL supernatant into 2mL microcentrifuge tube and spin at 10,000 rpm for 100 min. Take 1.00mL supernatant into 15mL centrifuge tube and add 9 mL 2% HNO3 for ICP-MS.

2nd sampling: 5/29/07 (two-day sorption) Centrifuge samples in 45mL test tube @ 3420 rpm for 10 min. Transfer 1.4 mL supernatant into 2mL microcentrifuge tube and spin at 10,000 rpm for 100 min. Take 1.00mL supernatant into 15mL centrifuge tube and add 9 mL 2% HNO3 for ICP-MS.

Appendix D: U Sorption Data Table

		Date	4/17/2007	4/17/2007	4/23/2007	4/24/2007		4/25/2007	4/25/2007	4/25/2007	4/25/2007	4/25/2007		5/25/2007			
S#	RN	Sample ID	solid mass, g	Water mass, g	pH before U spike	spike 241Am, g	Initial dpm/g	total volume prior to 1st sampling	pH @ 1st sampling	1st sampling dpm/g	**%sorbed U @ 1st sampling**	**Kd**	total volume prior to 2nd sampling	pH @ 2nd sampling	2nd sampling dpm/g	**%sorbed U @ 2nd sampling**	**Kd**
0	^{233}U	^{233}U in 2% HNO3 solution		45.2553		0.1003	126.7		Average dpm/g	127.5							
		^{233}U in 2% HNO3 solution							127.4	127.3							
		^{233}U in 2% HNO3 solution centrifuged								126.8							
		^{233}U in 2% HNO3 solution centrifuged							126.6	126.3					120.97		
										127.0					123.85		
1	^{233}U	ER-12-3 Blank		44.9726	8.06	0.0964	122.6	45.08	8.06	121.6	**0.8%**		43.28	7.86	116.59	**4.9%**	
4	^{233}U	ER-12-3 UE12t #2-1	3.0407	44.9501	8.20	0.1002	127.5	45.06	7.93	97.8	**23.3%**	**4.5**	43.26	7.90	49.99	**60.8%**	**23.0**
5	^{233}U	ER-12-3 UE12t #2-2	3.019	44.9634	8.23	0.1007	128.1	45.07	8.00	100.7	**21.4%**	**4.1**	43.27	7.91	51.57	**59.7%**	**22.1**
10	^{233}U	ER-12-3 UE12t #4-1	3.0349	44.9953	7.85	0.1021	129.7	45.11	7.69	73.5	**43.3%**	**11.4**	43.31	7.73	43.93	**66.1%**	**29.0**
11	^{233}U	ER-12-3 UE12t #4-2	3.0284	45.0034	7.77	0.1011	128.5	45.11	7.65	75.1	**41.5%**	**10.6**	43.31	7.60	43.09	**66.5%**	**29.5**
24	^{233}U	ER-12-3-UE12n #8-1	3.0197	44.9403	8.10	0.1007	128.1	45.05	7.93	58.0	**54.7%**	**18.0**	43.25	7.74	28.19	**78.0%**	**52.9**
25	^{233}U	ER-12-3-UE12n #8-2	3.0307	44.9953	7.90	0.1013	128.7	45.11	7.85	57.8	**55.1%**	**18.3**	43.31	7.64	31.74	**75.3%**	**45.5**
16	^{233}U	ER-12-3-UE12n 15A-1	3.0252	45.0052	7.78	0.101	128.3	45.12	7.72	68.2	**46.9%**	**13.1**	43.32	7.77	44.63	**65.2%**	**28.0**
17	^{233}U	ER-12-3-UE12n 15A-2	3.0046	45.2154	7.75	0.1012	128.0	45.33	7.68	66.2	**48.3%**	**14.1**	43.53	7.69	44.47	**65.3%**	**28.3**
2	^{233}U	T-Plug Blank		44.6159	8.71	0.1005	128.8	44.73	8.55	129.6	**0.0%**		42.93	8.44	124.80	**3.1%**	
6	^{233}U	T-Plug-UE12t #2-1	3.0604	44.9895	8.58	0.1006	127.9	45.10	8.48	129.4	**0.0%**	**N/A**	43.30	8.45	121.50	**5.0%**	**0.8**
7	^{233}U	T-Plug-UE12t #2-2	3.0156	45.0612	8.53	0.101	128.2	45.17	8.45	128.4	**0.0%**	**N/A**	43.37	8.37	120.18	**6.2%**	**1.0**
12	^{233}U	T-Plug-UE12t #4-1	3.0239	44.9445	8.25	0.1004	127.7	45.05	8.09	129.7	**0.0%**	**N/A**	43.25	8.11	119.77	**6.2%**	**1.0**
13	^{233}U	T-Plug-UE12t #4-2	3.0098	45.1051	8.25	0.0995	126.1	45.22	8.11	129.1	**0.0%**	**N/A**	43.42	8.02	119.19	**5.5%**	**0.9**
3	^{233}U	T-Plug ACT2 Blank		44.9502	8.40	0.1018	129.5	45.06	8.31	130.0	**0.0%**		43.26	8.26	124.57	**3.8%**	
8	^{233}U	T-Plug ACT2-UE12t #2-1	3.0162	44.8902	8.55	0.1012	128.9	45.00	8.25	127.5	**1.1%**	**0.2**	43.20	8.13	116.18	**9.9%**	**1.6**
9	^{233}U	T-Plug ACT2-UE12t #2-2	3.0213	44.8427	8.40	0.1003	127.9	44.95	8.19	127.6	**0.2%**	**0.0**	43.15	8.11	115.31	**9.8%**	**1.6**
14	^{233}U	T-Plug ACT2-UE12t #4-1	3.0417	45.0033	8.15	0.1015	129.0	45.11	8.08	126.7	**1.8%**	**0.3**	43.31	8.01	117.03	**9.3%**	**1.5**
15	^{233}U	T-Plug ACT2-UE12t #4-2	3.0167	44.888	8.05	0.0996	126.9	45.00	8.02	126.9	**0.0%**	**0.0**	43.20	7.96	115.83	**8.7%**	**1.4**
18	^{233}U	N-Main Blank		44.8242	8.62	0.1015	129.5	44.93	8.53	130.1	**0.0%**		43.13	8.47	125.05	**3.4%**	
20	^{233}U	N-Main-UE12n 15A-1	3.0163	45.131	8.10	0.0972	123.2	45.24	8.11	109.5	**11.1%**	**1.9**	43.44	8.13	94.18	**23.5%**	**4.6**
21	^{233}U	N-Main-UE12n 15A-2	3.0203	44.9519	8.12	0.1006	128.0	45.06	8.05	115.1	**10.0%**	**1.7**	43.26	8.09	97.69	**23.7%**	**4.6**
26	^{233}U	N-Main-UE12n #8-1	3.0166	45.0412	8.35	0.1005	127.6	45.15	8.26	118.0	**7.5%**	**1.2**	43.35	8.18	103.05	**19.2%**	**3.6**
27	^{233}U	N-Main-UE12n #8-2	3.0237	44.9458	8.35	0.1008	128.2	45.06	8.26	117.9	**8.1%**	**1.3**	43.26	8.15	105.39	**17.8%**	**3.2**
19	^{233}U	N-Main ACT2-2 Blank		44.926	8.25	0.0996	126.8	45.04	8.31	127.4	**0.0%**		43.24	8.35	121.34	**4.3%**	
22	^{233}U	N-Main ACT2-2-UE12n 15A-1	3.0256	44.9313	7.90	0.1003	127.6	45.04	7.95	111.1	**13.0%**	**2.2**	43.24	8.01	92.40	**27.6%**	**5.7**
23	^{233}U	N-Main ACT2-2-UE12n 15A-2	3.0136	44.9233	7.93	0.1014	129.1	45.03	7.97	114.8	**11.1%**	**1.9**	43.23	8.01	93.82	**27.3%**	**5.6**
28	^{233}U	N-Main ACT2-2-UE12n #8-1	3.0071	44.934	8.24	0.098	124.7	45.04	8.20	112.6	**9.7%**	**1.6**	43.24	8.10	98.12	**21.3%**	**4.1**
29	^{233}U	N-Main ACT2-2-UE12n #8-2	3.0161	44.914	8.25	0.1006	128.1	45.02	8.11	114.8	**10.4%**	**1.7**	43.22	8.07	103.15	**19.5%**	**3.6**

Sample list, 45 mL of water and 3g gram of solid per sample
1st sampling: 4/25/07 (one day sorption) Centrifuge samples in 45mL test tube @ 3420 rpm for 10 min. Transfer 1.8 mL supernatant into 2mL microcentrifuge tube and spin at 10,000 rpm for 100 min. Take 1.5mL supernatant for LSC.
2nd sampling: 5/25/07 (31 day sorption) Centrifuge samples in 45mL test tube @ 3220 rpm for 10 min. Transfer 1.7 mL supernatant into 2mL microcentrifuge tube and spin at 10,000 rpm for 100 min. Take 1.5mL supernatant for LSC.